ENT COOPERATION TR

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

Assistant Commissioner for Patents
United States Patent and Trademark Office
Box PCT
Washington, D.C.20231
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 06 June 2000 (06.06.00)	
International application No. PCT/GB99/03269	**Applicant's or agent's file reference** PFC 1430 PCT
International filing date (day/month/year) 04 October 1999 (04.10.99)	**Priority date** (day/month/year) 16 October 1998 (16.10.98)
Applicant GASCOYNE, John, Malcolm et al	

1. The designated Office is hereby notified of its election made:

   [X] in the demand filed with the International Preliminary Examining Authority on:

   10 May 2000 (10.05.00)

   [ ] in a notice effecting later election filed with the International Bureau on:

2. The election [X] was [ ] was not

   made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO **34, chemin des Colombettes** **1211 Geneva 20, Switzerland** Facsimile No.: (41-22) 740.14.35	Authorized officer S. Mafla Telephone No.: (41-22) 338.83.38

09/807 68

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PATENT COOPERATION TREATY

REC'D 24 OCT 2000

WIPO PCT

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PFC 1430 PCT	**FOR FURTHER ACTION**	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/GB99/03269	International filing date *(day/month/year)* 04/10/1999	Priority date *(day/month/year)* 16/10/1998
International Patent Classification (IPC) or national classification and IPC H01M8/02		
Applicant JOHNSON MATTHEY PUBLIC LIMITED COMPANY et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   I ☒ Basis of the report
   II ☐ Priority
   III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   IV ☐ Lack of unity of invention
   V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   VI ☒ Certain documents cited
   VII ☐ Certain defects in the international application
   VIII ☒ Certain observations on the international application

Date of submission of the demand 10/05/2000	Date of completion of this report 20.10.2000
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Mizera, E Telephone No. +49 89 2399 8580

Form PCT/IPEA/409 (cover sheet) (January 1994)

I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

Description, pages:

1-13	as originally filed

Claims, No.:

1-23	as originally filed

2. The amendments have resulted in the cancellation of:

☐	the description,	pages:
☐	the claims,	Nos.:
☐	the drawings,	sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1-23
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-23
Industrial applicability (IA)	Yes:	Claims	1-23
	No:	Claims	

2. Citations and explanations

see separate sheet

VI. Certain documents cited

1. Certain published documents (Rule 70.10)

and / or

2. Non-written disclosures (Rule 70.9)

see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

AS TO BOX V:

1. The following documents are cited:

   D1: EP-A-0 791 974 (JOHNSON MATTHEY PLC) 27 August 1997 (1997-08-27)
   D2: EP-A-0 631 337 (TANAKA PRECIOUS METAL IND ;WATANABE MASAHIRO (JP)) 28 December 1994 (1994-12-28) cited in the application

2. Although the present invention seems to be directed to a method that requires a **full incorporation** of the supported catalyst into the membrane, in order to manufacture a self-humidifying membrane (see the object of the invention, p.4, l.18-20), claim 1 only requires the catalysed support to be 'combined' with an ion-conduction polymer composition.

3. Consequently all the processes, in which a supported catalyst is bonded to a polymer electrolyte membrane, e.g. by application of heat and pressure, anticipate the teachings of claim 1. Such a process is already described in D1, col.9, l.26-33. In this case the platinum catalyst containing face of the cathode is bonded to the membrane electrolyte face. The platinum catalyst is supported on carbon black, but the whole assembly of supported catalyst and carbon fiber mats can be seen as a single catalysed support. This fully anticipates the taechings of claims 1-23. These claims do not meet the requirements of Art.33(2) PCT.

4. It is remarked that D1 does not represent the only document that teaches such embodiments. Class C25B 11/20 of the IPC is directed to such products and contains numerous relevant documents.

5. Also D2 remains to be considered. This document has alredy been discussed in the application, and was critisised as a complex process involving multiple steps(see p.3). Claim 1, however, does not exclude further steps, as steps (a) and (b) are only 'comprised'. Reference is made in particular to col.8, l.11-18, according to which the catalysed metal oxide can be incorporated in the solid polymer electrolyte. This incorporation can proceed in accordance with the procedures described in this document, but a skilled man would be well aware of a series of other possibilities (see e.g. p.8, l.16-31 of the present application).

6. Both with respect to D1 and D2 claims 1-23 lack the requirements of Art.33(2) and (3) PCT. As to claims 20-23 it is explicitly mentioned that novelty and inventive step would have to be shown irrespective of the process used for its production.

AS TO BOX VI:

EP-A-0 875 524

AS TO BOX VIII:

1. On p.2, l.23 the word 'application' should be substituted by 'specification'.

2. The references to unpublished documents (see p.8, l.30, 31) should be deleted.

3. On p.4, l.30 the 'incorporation bvy reference' should be deleted under Art.6 PCT.

4. On p.11 the SI unit should be added to the unit 'psi'.

PATENT COOPERATION TREATY

09/807682

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

WISHART, Ian Carmichael
Johnson Matthey Technology Centre
Blounts Court
Sonning Common
Reading RG4 9NH
GRANDE BRETAGNE

RECEIVED
24 OCT 2000

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

	Date of mailing *(day/month/year)* 20.10.2000
Applicant's or agent's file reference PFC 1430 PCT	**IMPORTANT NOTIFICATION**

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB99/03269	04/10/1999	16/10/1998

Applicant
JOHNSON MATTHEY PUBLIC LIMITED COMPANY et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

   The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

   Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

   For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	DA ROCHA, O. Tel.+49 89 2399-8101

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
PFC 1430 PCT		
International application No. PCT/GB99/03269	International filing date (*day/month/year*) 04/10/1999	Priority date (*day/month/year*) 16/10/1998
International Patent Classification (IPC) or national classification and IPC H01M8/02		
Applicant JOHNSON MATTHEY PUBLIC LIMITED COMPANY et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   I ☒ Basis of the report
   II ☐ Priority
   III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   IV ☐ Lack of unity of invention
   V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   VI ☒ Certain documents cited
   VII ☐ Certain defects in the international application
   VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
10/05/2000	20.10.2000
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Mizera, E Telephone No. +49 89 2399 8580

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB99/03269

I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

Description, pages:

1-13 as originally filed

Claims, No.:

1-23 as originally filed

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1-23
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-23
Industrial applicability (IA)	Yes:	Claims	1-23
	No:	Claims	

2. Citations and explanations

   see separate sheet

VI. Certain documents cited

1. Certain published documents (Rule 70.10)

   and / or

2. Non-written disclosures (Rule 70.9)

   see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

AS TO BOX V:

1. The following documents are cited:

   D1: EP-A-0 791 974 (JOHNSON MATTHEY PLC) 27 August 1997 (1997-08-27)
   D2: EP-A-0 631 337 (TANAKA PRECIOUS METAL IND ;WATANABE MASAHIRO (JP)) 28 December 1994 (1994-12-28) cited in the application

2. Although the present invention seems to be directed to a method that requires a **full incorporation** of the supported catalyst into the membrane, in order to manufacture a self-humidifying membrane (see the object of the invention, p.4, l.18-20), claim 1 only requires the catalysed support to be 'combined' with an ion-conduction polymer composition.

3. Consequently all the processes, in which a supported catalyst is bonded to a polymer electrolyte membrane, e.g. by application of heat and pressure, anticipate the teachings of claim 1. Such a process is already described in D1, col.9, l.26-33. In this case the platinum catalyst containing face of the cathode is bonded to the membrane electrolyte face. The platinum catalyst is supported on carbon black, but the whole assembly of supported catalyst and carbon fiber mats can be seen as a single catalysed support. This fully anticipates the taechings of claims 1-23. These claims do not meet the requirements of Art.33(2) PCT.

4. It is remarked that D1 does not represent the only document that teaches such embodiments. Class C25B 11/20 of the IPC is directed to such products and contains numerous relevant documents.

5. Also D2 remains to be considered. This document has alredy been discussed in the application, and was critisised as a complex process involving multiple steps(see p.3). Claim 1, however, does not exclude further steps, as steps (a) and (b) are only 'comprised'. Reference is made in particular to col.8, l.11-18, according to which the catalysed metal oxide can be incorporated in the solid polymer electrolyte. This incorporation can proceed in accordance with the procedures described in this document, but a skilled man would be well aware of a series of other possibilities (see e.g. p.8, l.16-31 of the present application).

6. Both with respect to D1 and D2 claims 1-23 lack the requirements of Art.33(2) and (3) PCT. As to claims 20-23 it is explicitly mentioned that novelty and inventive step would have to be shown irrespective of the process used for its production.

AS TO BOX VI:

EP-A-0 875 524

AS TO BOX VIII:

1. On p.2, l.23 the word 'application' should be substituted by 'specification'.

2. The references to unpublished documents (see p.8, l.30, 31) should be deleted.

3. On p.4, l.30 the 'incorporation bvy reference' should be deleted under Art.6 PCT.

4. On p.11 the SI unit should be added to the unit 'psi'.

PATENT COOPERATION TREATY

PCT

09/807682

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference PFC 1430 PCT	**FOR FURTHER ACTION** see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/GB 99/03269	International filing date *(day/month/year)* 04/10/1999	(Earliest) Priority Date *(day/month/year)* 16/10/1998
Applicant JOHNSON MATTHEY PUBLIC LIMITED COMPANY et al.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

- [X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**
   a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
      - [ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

   b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :
      - [ ] contained in the international application in written form.
      - [ ] filed together with the international application in computer readable form.
      - [ ] furnished subsequently to this Authority in written form.
      - [ ] furnished subsequently to this Authority in computer readble form.
      - [ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
      - [ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).
3. [ ] **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,
   - [ ] the text is approved as submitted by the applicant.
   - [X] the text has been established by this Authority to read as follows:

   PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE

5. With regard to the **abstract**,
   - [X] the text is approved as submitted by the applicant.
   - [ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. ______
   - [ ] as suggested by the applicant.
   - [ ] because the applicant failed to suggest a figure.
   - [ ] because this figure better characterizes the invention.
   - [ ] None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 99/03269

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01M8/02 C08J5/22

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01M C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 791 974 A (JOHNSON MATTHEY PLC) 27 August 1997 (1997-08-27)	1-7, 11-13, 16,17, 20-23
	column 4, line 25 - line 29; claims 1,3; example 2	
Y		1,14,15

P,X	EP 0 875 524 A (JOHNSON MATTHEY PLC) 4 November 1998 (1998-11-04) cited in the application	1,3,4, 11-13, 16,17, 20-23
	claims 14,18; example 1	

	-/--	

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
4 February 2000	11/02/2000

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Andrews, M

1

Form PCT/ISA/210 (second sheet) (July 1992)

INT[illegible]NATIONAL SEARCH REPORT

[illegible]ernational Application No
PCT/GB 99/03269

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 631 337 A (TANAKA PRECIOUS METAL IND ;WATANABE MASAHIRO (JP)) 28 December 1994 (1994-12-28) cited in the application claims 2,5,7 -----	1,14,15

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/GB 99/03269

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0791974 A	27-08-1997	AU 1473797 A	04-09-1997
		BR 9701086 A	24-11-1998
		CA 2198553 A	28-08-1997
		EP 0942482 A	15-09-1999
		JP 9326256 A	16-12-1997
		US 5865968 A	02-02-1999
EP 0875524 A	04-11-1998	JP 10312815 A	24-11-1998
EP 0631337 A	28-12-1994	JP 7090111 A	04-04-1995
		US 5766787 A	16-06-1998

PCT WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7 : H01M 8/02, C08J 5/22

A1

(11) International Publication Number: WO 00/24074

(43) International Publication Date: 27 April 2000 (27.04.00)

(21) International Application Number: PCT/GB99/03269

(22) International Filing Date: 4 October 1999 (04.10.99)

(30) Priority Data:
9822576.6 16 October 1998 (16.10.98) GB

(71) Applicant *(for all designated States except US)*: JOHNSON MATTHEY PUBLIC LIMITED COMPANY [GB/GB]; 2–4 Cockspur Street, Trafalgar Square, London SW1Y 5BQ (GB).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: GASCOYNE, John, Malcolm [GB/GB]; Lyduska, Routs Green, Bledlow Ridge, High Wycombe, Bucks HP14 4BB (GB). HARDS, Graham, Alan [GB/GB]; Chestnut Cottage, Tokers Green Lane, Tokers Green, Reading RG4 9EB (GB). HOGARTH, Martin, Philip [GB/GB]; 51 Orchard Avenue, Sonning Common, Reading RG4 9LT (GB). RALPH, Thomas, Robertson [GB/GB]; 94 Shaftesbury Road, Reading, Berkshire RG30 2QJ (GB).

(74) Agent: WISHART, Ian, Carmichael; Johnson Matthey Technology Centre, Blounts Court, Sonning Common, Reading RG4 9NH (GB).

(81) Designated States: CA, JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
With international search report.

(54) Title: PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE

(57) Abstract

A process for preparing a solid polymer electrolyte membrane comprising an ion–conducting polymer, a catalyst and a high surface area support material, which process comprises: (a) associating the catalyst with the support material to form a catalysed support; and (b) combining the catalysed support with an ion–conducting polymer composition; is disclosed. Also disclosed is a membrane electrode assembly and a fuel cell comprising a solid polymer electrolyte membrane prepared by the process of the invention.

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
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BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
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CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon			PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

PROCESS FOR PREPARING A SOLID POLYMER ELECTROLYTE MEMBRANE

The present invention relates to a process for the preparation of a solid polymer electrolyte membrane that is of use in electrochemical devices, particularly fuel cells.

Electrochemical cells invariably comprise an ion-conducting electrolyte located between two electrodes, the anode and cathode, at which the desired electrochemical reactions take place. Electrochemical cells may be found in a range of devices, for example fuel cells, batteries, sensors, electrodialysis reactors and electrolytic reactors. They have a diverse range of applications, including the electrolysis of water, chemical synthesis, salt splitting, water purification, effluent treatment and metal finishing, among others.

A fuel cell is an energy conversion device that efficiently converts the stored chemical energy of its fuel and oxidant into electrical energy. It does so by combining either hydrogen, stored as a gas or methanol, stored as a liquid or a gas, with oxygen to generate electrical power. The hydrogen or methanol is oxidised at the anode and oxygen is reduced at the cathode. Both electrodes are of the gas diffusion type. The combined structure formed from the membrane and the two gas diffusion electrodes is known as the membrane electrode assembly (MEA). An electric current is produced by the electron transfer reaction on the electrodes, with the flow of ions in the electrolyte completing the electrical circuit between the electrodes. The electrolyte therefore has to be in contact with both electrodes, and may be acidic or alkaline, and liquid or solid, in nature. In proton exchange membrane fuel cells (PEMFC), the electrolyte is a solid, ion-conducting, *i.e.* a proton-conducting, polymer membrane. The movement of ions between the electrodes is dependent upon the presence of water in the membrane. The membrane must be fully humidified to provide the efficient movement of ions *i.e.* maintain the highest possible specific conductivity. The membrane also prevents large quantities of hydrogen gas supplied at the anode and oxygen supplied at the cathode from passing through the electrolyte and mixing, although there is some transport of the reactant gases - a phenomenon referred to as "cross-over"- which leads to a reduction in cell performance. It is important that this cross-over is minimised. This is a particular problem with conventionally employed membranes at thicknesses below around 40μm, which are otherwise favoured because of their high conductivity in the fuel cell.

Conventionally, the membrane electrolytes useful in fuel cells are selected from commercially available membranes, for example the perfluorinated membranes sold under the trade names Nafion® (E I DuPont de Nemours and Co), Aciplex® (Asahi Chemical Industry) and Flemion® (Asahi Glass KK). In PEMFCs, the membrane electrolyte films - such as the Nafion® types of DuPont - have to be maintained in a highly water-saturated form in order to maximise their proton conductivity properties. Maintaining this high level of water content under operating conditions requires a complex water management system.

Attempts to control the humidity of the membrane have been made, which include saturating the hydrogen gas supplied to the anode and air supplied to the cathode with water vapour. However, this method brings with it its own disadvantages: it makes the overall system more complicated; lowers the fuel cell system efficiency; and can also reduce the cell's performance, particularly at high current densities, due to increased reactant gas (particularly oxygen in air) diffusional problems as a result of the increased amount of water in the electrode layers.

It has been recognised that one way to overcome these difficulties is to employ the concept of a self-humidified membrane. One way that has been suggested that this can be achieved is through the use of water-retaining particles such as highly-dispersed silica (SiO_2) incorporated into the membrane electrolyte film. The membrane can retain a higher level of water and is able to function with reduced need for humidifying the inlet gases.

It has also been discovered (European patent application EP 0631337) that it is possible to incorporate catalyst materials, such as those based on platinum, into the particle containing membrane. This further improves the water retaining ability of the membrane by reacting any hydrogen and oxygen that diffuse into the membrane from each of the catalysed electrode layers to produce water *in situ*. This also can permit the use of thinner membranes, with higher conductivity, by reducing the deleterious effects of reactant gas cross-over that would otherwise occur. Furthermore, the ability to use thinner membranes itself will further reduce the requirements for external humidification of the reactant gases. The back diffusion of water from cathode to anode is enhanced, thereby preventing the anode face of the membrane from drying.

The disclosure of EP 0631337 relates to a solid polymer electrolyte composition comprising solid polymer electrolyte and 0.01 to 80% (based on weight of electrolyte) of at least one metal catalyst, and the use of this composition in fuel cells. The specification further discloses the optional incorporation of 0.01 to 50% of particles and/or fibres of one or more metal oxides into the electrolyte. However, the method disclosed for making such compositions is multi-step and also has associated disadvantages. For example, one such composition is prepared by mixing a 5% w/w isopropanol solution of the electrolyte and an isopropanol dispersion of particles and/or fibres of 0.01 to 50% silica or titania and sufficiently agitating the mixture in an ultrasonic homogeniser. After this, the solution is transferred into a membrane moulding vessel and dried at room temperature, then vacuum-dried at 60°C to remove the isopropanol and form the membrane. To incorporate the metal catalyst, in the case of platinum, the solid polymer electrolyte membrane containing the metal oxide is dipped in distilled water containing a platinum-ammine complex, followed by agitation at 60°C for 5 hours, during which the platinum complex replaces the protons in the membrane by ion-exchange. After washing the membrane in distilled water for over 4 hours, it is dipped in distilled water containing a twenty times equivalence of hydrazine as reductant. After agitation at 60°C for another 5 hours the platinum particles are deposited in the solid polymer electrolyte containing the metal oxide. Then, after the ion-exchange group is protonised by means of treatment with 4 molar concentration hydrochloric acid, the membrane is washed and dried.

Hence, it can be seen that the prior art process for forming the material is complex and involves multiple steps, many of which are time-consuming and may not be easily capable of producing a consistent material. These factors increase the cost of producing this self-humidified membrane. It is essential for the future commercialisation of PEMFC power generation systems that the cost of the materials employed should be minimised. The polymer electrolyte membrane is one of the most expensive materials employed in the PEM fuel cell stack, due principally to the nature of the polymer itself, and its cost is currently too high to enable widespread commercialisation. Therefore, any process for producing more complex membranes needs to be simple, straightforward and low-cost.

A further expensive material in the PEM fuel cell stack is the platinum catalyst conventionally employed in the anode and cathode catalyst layers to promote the fuel cell electrode reactions of hydrogen oxidation and oxygen reduction. For many applications, particularly transportation, there is a need to minimise the use of these materials. It is widely believed that, for successful commercial exploitation, the level of platinum in the fuel cell stack should be reduced to a cell loading of as low as 0.20mg/cm^2 of the MEA. Therefore, whilst the incorporation of an additional amount of platinum catalyst into the membrane to effect the combination of any reactant gases permeating into the membrane to produce an improved self-humidified membrane is a worthwhile objective, it is a requirement that this additional amount of platinum be as low as possible. EP 0631337 and J. Electrochem. Soc. 143 (12) 3847-52 (1996) indicate that platinum loadings of 0.07mg/cm^2 are employed in the films produced by the above described multi-step process. At some 35% of the total platinum content that may be allowable for vehicular applications of the PEM fuel cell, this may be too high a loading for economic reasons. To our knowledge, therefore, membranes of this type have not yet found application in any of the current, practical pre-production demonstrations of PEM fuel cell power generators.

It is therefore an object of the present invention to overcome the disadvantages of the conventional ion-exchange membranes, without incurring the disadvantages of the prior art process for manufacturing a self-humidifying membrane.

Accordingly, the present invention provides a process for preparing a solid polymer electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high surface area support material, which process comprises:

(a) associating the catalyst with the support material to form a catalysed support; and

(b) combining the catalysed support with an ion-conducting polymer composition.

Preferably, the ion-conducting polymer composition is an aqueous ion-conducting polymer composition, for example as disclosed in European patent specification number EP 0731520, the contents of which is incorporated herein by reference in their entirety. Accordingly, the process of the present invention preferably comprises, as step (b), combining

the catalysed particulate material with an ion-conducting polymer in a liquid medium that is aqueous-based and is essentially free from organic solvents.

The process according to this invention, compared with the prior art multi-step process, is less complex, has fewer and shorter process steps, and is capable of producing a consistent and reproducible product; hence, the cost of producing the membrane is thereby reduced. Furthermore, as the catalyst is directly applied to the high surface area particulate material, which thereby acts as the support for the catalyst, the catalyst is also deposited in a high surface area, small particle size form. This enables the expensive catalyst to be used more effectively and in lower amounts whilst also improving the self-humidifying performance of the membrane. Additionally, when the particulate material has water-retaining properties, all of the catalyst promoting the recombination of hydrogen and oxygen is in immediate and intimate contact with the sites at which the water formed can be stored.

The catalyst may comprise any catalyst material capable of enabling the reaction of hydrogen and oxygen to form water to occur at PEM fuel cell temperatures of typically below 150°C. These include the precious metals, platinum, palladium or rhodium, or combinations thereof, or other transition group metals such as nickel or cobalt. Preferably, the precious metals platinum, palladium or rhodium, or combinations thereof are employed and, more preferably, platinum is used. Suitable catalysation processes, as already practised on a commercial scale, can be employed to form the catalyst material. These processes permit very close control of the amount of catalyst metal deposited onto the support, and thus subsequently incorporated into the membrane. Typically the catalyst metal can be deposited onto the support to loadings of between 0.01 to 50.0% by weight of the total catalysed support, and more preferably from 1 to 25 wt%, such as, for example 1-10 wt%. The amount of catalysed support incorporated into the membrane is such that the metal loading on the membrane is lower than $0.1 mg/cm^2$, preferably lower than $0.05 mg/cm^2$ and, more preferably, lower than $0.03 mg/cm^2$.

The high surface area support material may comprise any known, non-electrically conducting material which can used as a support for a catalyst, such as silica, titania, alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and zeolites. It may also be

advantageous to employ support materials that are known to retain high levels of water once suitably wetted, such as, for example, forms of silica. The material may be in the form of either fibres or small particles, such as particles with a mean particle size in the range of from 0.001μm to 10μm, preferably 0.01μm to 5μm.

For PEM fuel cell applications, the ion-conducting polymer is a proton-conducting polymer, examples of such polymers being well known to those skilled in the art. More than one proton-conducting polymer may be present and/or a non-ion-conducting polymer may also be included in the membrane prepared by the process of the invention.

The proton conducting polymers suitable for use in the present invention may include, but are not limited to:

1) Polymers which have structures with a substantially fluorinated carbon chain optionally having attached to it side chains that are substantially fluorinated. These polymers contain sulphonic acid groups or derivatives of sulphonic acid groups, carboxylic acid groups or derivatives of carboxylic acid groups, phosphonic acid groups or derivatives of phosphonic acid groups, phosphoric acid groups or derivatives of phosphoric acid groups and/or mixtures of these groups. Perfluorinated polymers include Nafion®, Flemion® and Aciplex® commercially available from E. I. DuPont de Nemours (U.S. Patents 3,282,875; 4,329,435; 4,330,654; 4,358,545; 4,417,969; 4,610,762; 4,433,082 and 5,094,995), Asahi Glass KK and Asahi Chemical Industry respectively. Other polymers include those covered in U.S. Patent 5,595,676 (Imperial Chemical Industries plc) and U.S. Patent 4,940,525 (Dow Chemical Co.)

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581, WO 95/08581 and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H, PO_2H_2, PO_3H_2, $CH_2PO_3H_2$, COOH, OSO_3H, OPO_2H_2, OPO_3H_2. Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

3) Fluorinated polymers such as those disclosed in EP 0 331 321 and EP 0345 964

(Imperial Chemical Industries plc) containing a polymeric chain with pendant saturated cyclic groups and at least one ion exchange group which is linked to the polymeric chain through the cyclic group.

4) Aromatic polymers such as those disclosed in EP 0 574 791 and US Patent 5,438,082 (Hoechst AG) for example sulphonated polyaryletherketone. Also aromatic polymers such as polyether sulphones which can be chemically grafted with a polymer with ion exchange functionality such as those disclosed in WO 94/16002 (Allied Signal Inc.).

5) Nonfluorinated polymers include those disclosed in U.S. Patent 5,468,574 (Dais Corporation) for example hydrocarbons such as styrene-(ethylene-butylene)- styrene, styrene-(ethylene-propylene)-styrene and acrylonitrile-butadiene-styrene co- and terpolymers where the styrene components are functionalised with sulphonate, phosphoric and/or phosphonic groups.

6) Nitrogen containing polymers including those disclosed in U.S. Patent 5,599,639 (Hoechst Celanese Corporation), for example, polybenzimidazole alkyl sulphonic acid and polybenzimidazole alkyl or aryl phosphonate.

7) Any of the above polymers which have the ion exchange group replaced with a sulphonyl chloride (SO_2Cl) or sulphonyl fluoride (SO_2F) group rendering the polymers melt processable. The sulphonyl fluoride polymers may form part of the precursors to the ion exchange membrane or may be arrived at by subsequent modification of the ion exchange membrane. The sulphonyl halide moieties can be converted to a sulphonic acid using conventional techniques such as, for example, hydrolysis.

Non-ion conducting polymeric materials which may be used in addition to the one or more ion conducting or proton conducting polymers include PTFE, FEP, PVDF, Viton® and hydrocarbon types such as polyethylene, polypropylene and polymethylmethacralate.

Other ion-conducting polymeric materials which are not proton conducting polymers may be used. For example, such polymers can be used for applications requiring an anion exchange membrane. Anion exchange polymers are generally based on quaternary ammonium groups, rather than the fixed sulphonic acid groups in proton conducting polymers. These include, for example, the tetraalkyl ammonium group ($-N^+R_3$) and the quaternary ammonium centre in Tosflex® membranes ($-N(R_1)(CH_2)_yN^+(R_3)$) supplied by Tosoh. However, it can be

envisaged that all of the proton exchange polymers described above could have anion exchange equivalents.

Most preferably, the ion-conducting polymer is an essentially aqueous solution of a perfluorinated co-polymer with ion-exchange groups, such as Nafion® (as mentioned hereinbefore).

The solid polymer electrolyte membrane prepared by the process of the present invention may be employed as or as a component of the ion-exchange membrane of a solid polymer electrolyte chemical cell, such as a PEMFC. Accordingly, the present invention further provides a membrane prepared by the process of the present invention. The membrane prepared by the process of the present invention is suitably for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200μm and preferably less than 100μm.

The catalysed support can be incorporated into the membrane by a number of methods:

The catalysed support, when in particle or fibre form, can be added directly to an solution of the proton-conducting polymer electrolyte material; preferably, an essentially aqueous solution as described in EP 0731520; the use of an aqueous solution permits the direct addition of the pre-formed catalysed support to the polymer solution, and eliminates the undesirable risk of combustion of the organic solvents that are present in the typically-employed polymer solutions. It is then possible to cast a membrane directly from this mixture according to any of the commonly practised methods in the art to produce a conventional unreinforced membrane. Alternatively, the mixture can be used to form a composite membrane by applying it to a pre-formed reinforcing substrate material such as disclosed in the art, for example an expanded PTFE matrix as disclosed in US 5,599,614 (Bahar et al) or a woven or non-woven fibre network of, for example, glass, silica or polymer fibres as disclosed for example in EP 0875524, unpublished UK Patent Application No 9822569.1 or unpublished UK Patent Application No 9822571.7.

Alternatively, when used in a composite membrane, the catalysed support may, when in particle form, itself comprise the binder used to form the fibre network. Subsequent to the formation of the fibre network or matrix comprising the catalysed support as a binder, a proton-conducting polymer is coated onto the network to produce the final membrane. Alternatively, when in fibre form, the catalysed support may itself form the fibre network prior to coating with a binder and subsequently coating with proton-conducting polymer to form the membrane.

In a further embodiment, a laminated membrane comprising more than one polymer-containing layer is provided, at least one layer of which is a membrane prepared by the process of the invention. Using such a laminated structure, it is possible, for example, to tailor the properties of the laminate membrane opposed to the anode and cathode sides in the MEA of a proton exchange membrane fuel cell, for example, to improve water management in the fuel cell, or to be able to use lower cost proton-conducting polymers to form a substantial part of the laminate membrane.

The present invention also relates to an MEA and a method for the manufacture thereof, wherein the MEA comprises a membrane produced by a process according to the present invention. A further aspect of the present invention relates to a fuel cell and a method for the manufacture thereof, which fuel cell comprises such a membrane and/or MEA.

The present invention will now be described by way of the following non-limiting example(s).

EXAMPLE 1

Catalyst Preparation

A 5.6 wt% Pt catalyst supported on silica (Davisil™, grade 644 from Aldrich, The Old Brickyard, New Road, Gillingham, Dorset SP8 4XT) was prepared. The silica (50g) was dispersed in 5 moldm^{-3} nitric acid to clean it and the mixture filtered. The silica filter cake was washed with demineralised water, dried and calcined in air at 500°C for 2 hours. The Pt salt, $Pt(NH_3)_4(OH)_2$ (28.87g), was placed in a beaker and sufficient demineralised water added to

produce 100g of solution. The Pt salt solution was added to the calcined silica and the mixture stirred for 5 minutes to allow the silica to adsorb the Pt salt solution. The resultant thick slurry was dried overnight at 60°C in a vacuum oven. It was then calcined in air at 300°C for 2 hours. The resultant 5.6 wt% Pt catalyst supported on silica showed a high level of Pt dispersion on the silica support as shown by a CO metal area of 111m^2/gPt and in transmission electron micrographs 1 to 3nm Pt particles, with some clustering forming collections of Pt particles 25nm across.

Preparation of Mixed Amorphous Silica Substrate

A mixture of chopped silica fibres (0.08g of type QC9/33 - 20mm from Quartz et Silice BP 521-77794 Nemours, Cedex, France and 0.47g of Q fibre type 104 from Johns Manville, Insulation Group, PO Box 5108, Denver, CO, USA) were dispersed with mixing in water (3000cm^3). A non-woven matrix was fabricated from the resulting mixture in a single-step process, based on the principles of paper-making technology, at a sheet size of 855cm^2 (33cm diameter) in a sheet former (design based on standard SCA sheet former from AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The fibre sheet was removed from the wire and air dried at 150°C. The non-woven sheet was sprayed with Nafion® 1100 EW solution (5wt% solution in lower aliphatic alcohols from Solution Technologies Inc., Mendenhall, PA 19357, USA) to bind the fibres together. This gives a dry Nafion loading of 0.29mg/cm^2.

Formation of Composite Membrane

The mixed amorphous silica substrate was first cast with aqueous Nafion® solution (50g, 5wt% solution), prepared as described in patent specification numbers EPA 731 520 and US 5,716,437, to produce, after drying, a substrate with a Nafion® layer applied to and partially embedded within one face of the substrate. Aqueous Nafion® solution (12g, 10.3wt%) was heated to 80°C and the 5.6 wt% Pt on silica catalyst (0.28g) added and the mixture silversoned at medium speed for 10 minutes. The mixture was then sprayed onto the second face of the substrate to produce a thin layer of Pt catalyst and dried in the oven at 80°C. This step was repeated until all of the mixture was sprayed onto the second face of the substrate, which was then dried at 150°C to produce a membrane. An identical membrane was

then prepared and the two membranes were laminated with the Pt catalyst face of each membrane facing each other at a temperature of 177°C and a pressure of 296psi for 6 minutes. This produced a composite membrane of thickness 58μm (as measured by electron probe microanalysis (EPMA)) with a central region that contained the Pt on silica catalyst at a Pt loading of $0.05mg/cm^2$ and with an overall Nafion® loading of $13.2mg/cm^2$.

COMPARATIVE EXAMPLE 1

An identical pair of membranes were prepared from sheets of mixed amorphous silica substrate as detailed above, but without the addition of the Pt on silica catalyst. The two membranes were laminated at a temperature of 177°C and a pressure of 296psi for 6 minutes to produce a composite membrane of comparable thickness (55μm) and Nafion® loading ($14.2mg/cm^2$) to the composite membrane containing the Pt on silica catalyst.

SINGLE CELL RESULTS

The membranes of Example 1 and Comparative Example 1 were formed into a membrane electrode assembly (MEA) using a carbon supported Pt/Ru catalyst containing anode ($0.25mgPt/cm^2$) and a carbon supported platinum catalyst containing cathode ($0.60mgPt/cm^2$). An evaluation of the MEA comprising the membrane was performed in a single cell operating at a temperature of 80°C with H_2/O_2 as reactants each at a pressure of 3 atmospheres absolute and reactant stoichiometries of 1.5 for H_2 and 10.0 for O_2.

A clear determination of the degree of H_2 cross-over from the anode to cathode of the MEA is provided by the open circuit voltage (OCV). The OCV is the cell potential when there is no net current flowing in the single cell and is determined by the difference between the cathode and anode potentials. Since hydrogen oxidation at the anode is extremely facile the anode potential is essentially constant in the MEAs examined herein and, consequently the OCV is a relative measure of the cathode potential. The cathode potential is a mixed potential which is determined by the electrode potentials for oxygen reduction ($\frac{1}{2}O_2 + 2H^+ + 2e^- = H_2O$), carbon corrosion ($C + H_2O = CO_2 + 2H^+ + 2e^-$) and hydrogen oxidation ($H_2 = 2H^+ + 2e^-$).

Since the change in the oxygen reduction and carbon corrosion potentials is negligible for the MEAs tested herein, any change in the OCV reflects a change in the concentration of hydrogen at the cathode i.e. reflects a change in the rate of hydrogen cross-over from anode to cathode through the membrane of the MEA.

Table 1 shows the OCV values of the MEAs based on the composite membrane of Example 1 at a platinum loading of 0.05mgP/cm^2 and the composite membrane of Comparative Example 1 that does not contain the Pt on silica catalyst. Also, shown for further comparison are the OCV values for identical MEAs based on commercially available Nafion® membranes (from E.I. DuPont de Nemours and Co.22828 NC 87 Hwy. W. Fayetteville, NC 28306). The OCV of the MEA based on the composite membrane of Example 1 is significantly higher than either the OCV of the MEA based on the composite membrane of Comparative Example 1 that does not contain the Pt on silica catalyst or the OCV values of the MEAs based on the Nafion® membranes. This increase in OCV reflects a decrease in the hydrogen concentration at the cathode reflecting the reduction in the rate of hydrogen cross-over due to the recombination of the H_2 with O_2 at the Pt on silica catalyst located within the composite membrane. The extent of the reduction in hydrogen cross-over can be calculated from the change in the OCV by using the Nernst Equation which shows,

$$(OCV^2 - OCV^1) = RT/nF \ln (p_{H2}{}^1 / p_{H2}{}^2)$$

where p_{H2} is the hydrogen pressure at the cathode. By using this equation the H_2 cross-over relative to Nafion® 117 can be calculated. As shown in Table 1 the rate of H_2 cross-over through the composite membrane of Example 1 is reduced by 85% compared to both Nafion® 117 and the composite membrane of Comparative Example 1, which does not contain the Pt on silica catalyst. This is despite the composite membrane of Example 1 being significantly thinner than Nafion® 117 in the fabricated MEAs (from EPMA measurements) as shown in Table 1. Further, as expected Nafion® 117 gives the lowest H_2 cross-over of the commercial membranes tested, since it is the thickest. This is shown in Table 1 by the H_2 cross-over of the commercial membranes relative to Nafion® 117, with all showing increased rates of H_2 cross-over with as expected, the progressively thinner Nafion® membranes showing progressively higher rates of H_2 cross-over relative to Nafion® 117. This highlights just how effective the composite membrane of Example 1 is at reducing H_2 cross-over by recombining

the H_2 with O_2 to produce water. This allows thinner membranes to be employed in the PEMFC, providing the benefits of both higher MEA performance and operation with reduced humidification, due to a higher water content in the membrane and the thinner membrane both promoting the rate of water back- diffusion from the cathode to anode.

Table 1

OCVs and reduction in H_2 cross-over due to the composite membrane containing the Pt on silica catalyst

Membrane	OCV / V	Membrane Thickness [a] / µm	H_2 cross-over Relative to Nafion® 117 / %
Example 1	1.058	58	- 85
Comparative Example 1	1.029	55	0
Nafion® 112	1.005	40	+79
Nafion® 1135	1.018	75	+52
Nafion® 115	1.027	100	+12
Nafion® 117	1.029	150	0

[a] From EPMA measurements

CLAIMS

1. A process for preparing a solid polymer electrolyte membrane comprising an ion-conducting polymer, a catalyst and a high surface area support material, which process comprises:
   (a) associating the catalyst with the support material to form a catalysed support; and
   (b) combining the catalysed support with an ion-conducting polymer composition.

2. A process according to claim 1, which comprises, as step (b), combining the catalysed support with an ion-conducting polymer in a liquid medium that is aqueous-based and is essentially free from organic solvents.

3. A process according to claim 1 or claim 2, wherein the catalyst comprises one or more precious metals, or combinations thereof, and/or other transition group metals.

4. A process according to any preceding claim, wherein the catalyst comprises platinum.

5. A process according to any preceding claim, wherein the catalyst is deposited onto the support material to a loading of between 0.01 to 50.0% by weight of the total catalysed support.

6. A process according to claim 5, wherein the catalyst is deposited onto the support material at a loading of from 1 to 25 wt% of the total catalysed support.

7. A process according to claim 6, wherein the catalyst is deposited onto the support material at a loading of from 1 to 10 wt% of the total catalysed support.

8. A process according to any preceding claim, wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than $0.1 mg/cm^2$.

9. A process according to claim 8 wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than $0.05mg/cm^2$.

10. A process according to claim 9 wherein the amount of catalysed support incorporated into the membrane is such that the metal loading is lower than $0.03mg/cm^2$.

11. A process according to any preceding claim, wherein the high surface support material is non-electrically conducting.

12. A process according to any preceding claim, wherein the high surface area support material is selected from silica, titania, alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and zeolites.

13. A process according to any preceding claim, wherein the support material is in the form of fibres.

14. A process according to any one of claims 1 to 12, wherein the support material is in the form of particles with a mean particle size in the range of from 0.001μm to 10μm.

15. A process according to claim 14, wherein the mean particle size is in the range of from 0.01μm to 5μm.

16. A process according to any preceding claim, wherein the ion-conducting polymer composition is an essentially aqueous solution of a perfluorinated co-polymer with ion-exchange groups.

17. A process according to any preceding claim, wherein the catalysed support, in particle or fibre form, is added directly to a solution of the ion-conducting polymer electrolyte.

18. A process according to any of claims 1 to 11 or 13 to 15, wherein the catalysed support is in particle form and is applied as a binder to form a fibre network to which the ion-

conducting polymer is subsequently applied to produce the membrane.

19. A process according to any of claims 1 to 13 or 16, wherein the catalysed support is in fibre form and itself is formed into a fibre network which is thereafter bound with a binder, and the ion-conducting polymer is subsequently applied to produce the membrane.

20. A membrane prepared by a process according to any preceding claim.

21. A membrane electrode assembly comprising a membrane prepared by a process according to any one of claims 1 to 19.

22. A fuel cell comprising a membrane prepared by a process according to any one of claims 1 to 19.

23. A fuel cell comprising a membrane electrode assembly according to claim 21.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 99/03269

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01M8/02 C08J5/22

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01M C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 791 974 A (JOHNSON MATTHEY PLC) 27 August 1997 (1997-08-27)	1-7, 11-13, 16,17, 20-23
	column 4, line 25 - line 29; claims 1,3; example 2	
Y		1,14,15

P,X	EP 0 875 524 A (JOHNSON MATTHEY PLC) 4 November 1998 (1998-11-04) cited in the application	1,3,4, 11-13, 16,17, 20-23
	claims 14,18; example 1	

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
4 February 2000	11/02/2000

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Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Intern...nal Application No
PCT/GB 99/03269

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 631 337 A (TANAKA PRECIOUS METAL IND ;WATANABE MASAHIRO (JP)) 28 December 1994 (1994-12-28) cited in the application claims 2,5,7 -----	1,14,15

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern ıal Application No
PCT/GB 99/03269

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